



Service life of metakaolin-based concrete exposed to carbonation Comparison with blended cement containing fly ash, blast furnace slag and limestone filler



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ABSTRACT

Carbonation of cement-based materials can lead to corrosion of the steel bars in reinforced concrete, and supplementary cementing materials (SCMs) often increase the carbonation kinetics compared to reference concretes made of cements composed of clinker only. The aim of this work was to assess the consequences, in terms of service life of structures, of a possible increase in the carbonation depth of metakaolin concretes. Experimental testing using accelerated (4% CO₂) and natural carbonation conditions showed that increasing the metakaolin content tended to increase the carbonation depth of concretes, due to the consumption of portlandite by the pozzolanic reaction. However, most of the time, the carbonation was within the range of carbonation depths found in commercially available blended cements (including fly ash, GGBS or limestone filler) that had already proved their worth on the market. The combination of MK and limestone filler (in CEM II/A-LL with 15% MK) behaved very well with respect to carbonation, the carbonation depth being almost equivalent to that of CEM I samples. The modelling of CO₂ ingress into the concretes showed that, although metakaolin increased the carbonation (except when associated with limestone filler), the carbonation depth did not exceed 30 mm after 50 years, far from the value of 50 mm generally used as concrete cover to protect the steel bars. This means that the formulations including metakaolin would not have deteriorated by the end of the building's service life.

1. Introduction

Carbonation of cement-based materials is an important pathology that can cause severe degradation of concrete, since it reduces the pH of the pore solution and thus leads to the corrosion of steel rebars through the destruction of their passivation layer [1]. This decrease of the pH is due to the consumption of portlandite (produced by the hydration of the cement) by atmospheric CO₂. The pozzolanic reaction of metakaolin (MK)—and more generally of all other supplementary cementing materials (SCM)—leads to a decrease of the portlandite reserve in the cement paste, so several authors have concluded [2–6] that the replacement of Portland cement by metakaolin usually causes an increase of the carbonation kinetics of concrete.

Carbonation results obtained by San Nicolas [2] after concrete exposure of 28 days in a carbonation chamber (50% CO₂, 20 °C and 65% relative humidity) showed that the carbonation depth increased from 2 to 9 mm (+ 450%) when 25% of cement was replaced by metakaolin. These results were in correlation with the work of Kim et al. [3], who observed increases of 40, 70, 100 and 370% for the replacement of 5, 10, 15 and 25% of cement, respectively, by

metakaolin (56 days at 5% CO₂, 30 °C and 60% relative humidity). McPollin et al. [4] confirmed these results with 10% of metakaolin (5% CO₂, 20 °C and 65% relative humidity). Mejía de Gutiérrez et al. [5] studied the effect of the duration of the wet cure of metakaolin-based mixtures on the carbonation kinetics (exposure for 3 and 6 weeks in a carbonation chamber: 2.25% CO₂, 30 °C and 70% of relative humidity), as it is well known that the pozzolanic reaction is slower than the cement hydration reaction. Their results were in agreement with those in the literature [2–4], with a greater carbonation depth in metakaolin concrete than in a reference sample when the curing time was limited to 28 days. However, for longer curing times (wet cure of 240 days), the carbonation kinetics was slower in the case of concrete made with metakaolin than in the control concrete with cement only.

The use of other SCMs also causes an increase in the carbonation kinetics when compared to a reference sample composed of cement only. In the case of ground granulated blast furnace slag (GGBS), high doses could lead to a significant increase in the carbonation depth [6,7]. This effect can be reduced by longer wet curing [8,9] or by the use of finer GGBS [9]. Increases in the carbonation kinetics are also seen when high doses of silica fume [10] or fly ash (> 30%) are used

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[11]. However, Bai et al. [12] showed that the combination of metakaolin and fly ash led to a reduction of the carbonation kinetics with respect to a concrete with fly ash only (4% of CO₂).

The aim of the present work was to assess the consequences of the possible increase in the carbonation depth of metakaolin concretes, in terms of service life of structures. For this purpose, three points were treated and are reported in this paper:

- 1- Most of the carbonation data found in the literature concerns relatively pure metakaolin and a rotary kiln. The work presented here used flash-calcined metakaolin having a particular composition [13]. The reported data could thus improve our knowledge on the effect of different types of metakaolin on the concrete carbonation process.
- 2- The most typical studies found in regard to carbonation of SCM matrices compare a pure Portland cement and an SCM concrete containing less cement, so less portlandite. The carbonation results are rarely in favor of the SCM, whatever its nature. Nevertheless, the use of blended cements remains the usual practice when the aim is to improve other durability aspects of concrete. These blended cements are accepted for carbonation exposure classes (governed by the XC class in European standard EN 206-1 [14]), even if they are used in concretes subjected to carbonation, as long as they are included in a standard such as EN 197-1 [15]. For this reason, several commercial blended cements (based on limestone filler, blast furnace slag and fly ash) were used as references and to provide a basis of comparison for accelerated and natural carbonation studies of metakaolin concretes.
- 3- Even in the case of an increase of the carbonation of concrete with SCMs, it remains pertinent to evaluate the possible consequences of such behavior in terms of service life of the concrete. A model [16] predicting the carbonation depth of reinforced concrete structures, adjusted with the experimental results of natural carbonation, was thus used to compare the long-term carbonation behavior of pure Portland cement, blended cements and metakaolin concretes.

2. Materials and methods

2.1. Materials

Table 1 gives the characteristics of the binders used in this study. All the cements were commercial ones and complied with the European standard EN 197-1 [15]. Two plain cements containing a minimum of 95% of clinker were used as references: normal (CEM I) and low-sulfate (CEM I PM-ES) cement, the latter containing less C₃A. Five blended cements containing one or a combination of additions were also used as references:

- Limestone cement: a CEM II/A-LL (16% limestone filler);
- Fly ash cement: a CEM II/A-V with 15% of fly ash;
- Blast furnace slag cement: a CEM III/A with 62% of GGBS;
- Combination of GGBS and fly ash cement: a CEM V/A with 22% of each addition.

The metakaolin used was a flash calcined one [17]. Flash calcination refers to the combustion process where the particles of kaolinite are transformed into metakaolin by passing near a flame (temperature around 700 °C) for a few tenths of a second [18]. This process is faster and consumes less energy than traditional methods (e.g. rotary kiln) [13]. Due to the lower purity of the deposit, this metakaolin has an impurity rate of about 50%, mainly composed of quartz. This leads to a marked decrease in the surface area and therefore in water demand compared to a pure metakaolin. The siliceous aggregates used were divided into six size classes (0–0.315 mm, 0.315–1 mm, 1–4 mm, 4–8 mm, 8–12 mm, 12–20 mm). The particle size distribution of the aggregates in the concretes was established by means of the Dreux method in order to optimize the compactness of the grading curve [18]. The High Range Water Reducing Agent (HRWRA) was a polycarboxylate type available as a commercial solution (density = 1.05 kg/m³, active solid content by weight = 30.5%).

2.2. Concrete design

Table 2 summarizes the 11 concrete designs evaluated in this study. Several concretes without metakaolin were cast to be used as references of commercial mixtures accepted in the ready-mixed concrete industry:

Table 1
Composition and physical properties of the commercial cements and metakaolin.

Cement type	CEM I	CEM I PM-ES	CEM III/A	CEM II/A-LL	CEM II/A-V	CEM V/A	Metakaolin
Additions	–	–	Blast furnace slag	Limestone filler	Fly ash	Blast furnace slag and Fly ash	–
Strength class	52.5	52.5	52.5	42.5	42.5	42.5	–
Chemical properties (%)							
SiO ₂	19.75	21.70	26.20	18.00	25.55	28.70	67.10
Al ₂ O ₃	5.27	3.70	7.44	3.80	8.08	10.00	26.80
CaO	63.97	65.70	55.00	63.00	55.54	46.80	1.12
MgO	1.93	1.00	4.00	1.20	1.02	2.60	0.11
Fe ₂ O ₃	2.39	4.60	1.70	2.20	3.44	3.40	2.56
K ₂ O	0.48	0.29	0.36	0.70	1.09	1.27	0.12
Na ₂ O	0.17	0.16	0.57	0.09	0.20	0.24	0.01
TiO ₂	–	0.23	–	0.20	0.40	0.60	1.37
SO ₃	2.95	2.59	1.82	2.60	2.55	2.80	–
Cl [–]	0.03	0.02	0.33	0.03	0.01	0.01	–
Loss on ignition	1.77	0.90	2.23	7.70	1.40	2.10	0.84
Physical properties							
Specific density (kg/m ³)	3140	3190	3010	3070	3000	2910	2510
Blaine/BET specific surface area (cm ² /g)	3801	3550	4300	4085	3613	4900	165 000 (BET)
Compressive strength MPa (28 days)	65.3	60.8	64.7	54.1	49.4	48.1	–
Bogue composition (clinker) (%)							
C ₃ S	58.1	64.0	58.1	65.0	63.0	66.0	–
C ₂ S	10.0	11.0	10.0	13.0	12.9	11.0	–
C ₃ A	10.7	2.5	10.7	8.0	7.9	7.0	–
C ₄ AF	8.0	14.1	10.0	8.0	9.7	11.0	–

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